



**PATENT APPLICATION**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of

Hirohisa Tanaka et al.

Appln. No.: 10/520,068

Group Art Unit: 1754

Filed: January 5, 2005

Examiner: Timothy C. VANOV

For: METHOD FOR PRODUCING PEROVSKITE-TYPE COMPOSITE OXIDE

**DRAFT DECLARATION UNDER 37 C.F.R. § 1.132**

United States Patent and Trademark Office  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, Virginia 22313-1450

Sir:

I, Kimiyoshi KANEKO, hereby declare and state:

THAT I am a citizen of Japan;

THAT I received a Degree from the Division of Chemistry in the Faculty of Science of  
Hokkaido University in March, 1969;

THAT I was employed by Hokko Chemical Industry Co., Ltd. from April, 1969, ultimately  
holding the position of Group Leader of the Fine Ceramics in the Fine Chemicals Development  
Division, and that I engaged in the development and production of the METHOD FOR  
PRODUCING PEROVSKITE-TYPE COMPOSITE OXIDE claimed in the present application;

THAT I became a co-inventor of the above-identified application;

THAT I have reviewed the Office Actions of June 5, 2007;

EXAMPLE 1

Preparation of Sr<sub>3</sub>NiPtO<sub>6</sub> (Pt content: 31.84 % by weight)

After toluene was distilled off from 55.34 g of toluene solution of strontium isopropoxide (0.0492 mol), a mixed solution containing 213 g of 2-methoxyethanol and 640 g of isopropyl alcohol was added thereto for dissolution. Subsequently, 6.450 g (0.0164 mol) of platinum acetylacetonate was added and the solution was heated at 70°C for three hours.

Then, 4.081 g (0.0164 mol) of nickel acetate tetrahydrate was added and the solution was heated at 70°C for one hour. After the heating, 1.77 g of deionized water was added and the solution was heated at 70°C for two hours.

Thereafter, the resulting mixture was separated from the solvent by distillation, dried in a vacuum at 100°C, and baked at 500°C for three hours or 1000°C for one hour to prepare Sr<sub>3</sub>NiPtO<sub>6</sub>.

EXAMPLE 2

Preparation of Sr<sub>4</sub>PtO<sub>6</sub> (Pt content: 30.41 % by weight)

After toluene was distilled off from 71.99 g of toluene solution of strontium isopropoxide (0.0640 mol), a mixed solution containing 208 g of 2-methoxyethanol and 624 g of isopropyl alcohol was added thereto for dissolution. Subsequently, 6.293 g (0.0160 mol) of platinum acetylacetonate was added and the solution was heated at 70°C for three hours.

Then, 1.77 g of deionized water was added and the solution was heated at 70°C for two hours.

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Thereafter, the resulting mixture was separated from the solvent by distillation, dried in a vacuum at 100°C, and baked at 500°C for three hours or 1000°C for one hour to prepare  $\text{Sr}_4\text{PtO}_6$ .

EXAMPLE 3

Preparation of  $\text{MgAl}_2\text{O}_4$  Supporting  $\text{Sr}_3\text{NiPtO}_6$  (Pt content: 1.25 % by weight)

After toluene was distilled off from 0.562 g of toluene solution of strontium isopropoxide (0.5 mmol), a mixed solution containing 11 g of 2-methoxyethanol and 33 g of isopropyl alcohol was added thereto for dissolution. Subsequently, 0.0657 g (0.167 mmol) of platinum acetylacetonate was added and the solution was heated at 70°C for three hours.

The obtained solution was added dropwise to a suspension prepared by mixing 2.50 g of  $\text{MgAl}_2\text{O}_4$  (specific surface area of 37  $\text{m}^2/\text{g}$ ), 11 g of 2-methoxyethanol and 33 g of isopropyl alcohol, and the suspension was heated at 70°C for one hour. Then, 0.0415 g (0.167 mmol) of nickel acetate tetrahydrate was added thereto and the suspension was heated at 70°C for one hour. After the heating, 0.018 g of deionized water was added and the suspension was heated at 70°C for two hours.

Thereafter, the resulting mixture was separated from the solvent by distillation, dried in a vacuum at 100°C, and baked at 500°C for three hours or 1000°C for one hour to prepare  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_3\text{NiPtO}_6$ .

#### EXAMPLE 4

##### Preparation of MgAl<sub>2</sub>O<sub>4</sub> Supporting Sr<sub>4</sub>PtO<sub>6</sub> (Pt content: 1.25 % by weight)

After toluene was distilled off from 0.751 g of toluene solution of strontium isopropoxide (0.668 mmol), a mixed solution containing 11 g of 2-methoxyethanol and 33 g of isopropyl alcohol was added thereto for dissolution. Subsequently, 0.0657 g (0.167 mmol) of platinum acetylacetonate was added and the solution was heated at 70°C for three hours.

The obtained solution was added dropwise to a suspension prepared by mixing 2.50 g of MgAl<sub>2</sub>O<sub>4</sub> (specific surface area of 37 m<sup>2</sup>/g), 11 g of 2-methoxyethanol and 33 g of isopropyl alcohol, and the suspension was heated at 70°C for one hour. Then, 0.024 g of deionized water was added and the suspension was heated at 70°C for two hours.

Thereafter, the resulting mixture was separated from the solvent by distillation, dried in a vacuum at 100°C, and baked at 500°C for three hours or 1000°C for one hour to prepare MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>4</sub>PtO<sub>6</sub>.

#### EVALUATION

##### Reduction/Oxidation Treatment

Each powder of Sr<sub>3</sub>NiPtO<sub>6</sub>, Sr<sub>4</sub>PtO<sub>6</sub>, MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>3</sub>NiPtO<sub>6</sub> and MgAl<sub>2</sub>O<sub>4</sub> supporting Sr<sub>4</sub>PtO<sub>6</sub> obtained above was subjected to a reduction treatment under a reductive atmosphere (CO: 7.5 %, H<sub>2</sub>: 2.5 %, CO<sub>2</sub>: 8.0 %, and N<sub>2</sub>: Balanced) at 800°C for 1 hour, and subsequently to an oxidation treatment under an atmospheric atmosphere at 800°C for one hour.

X-Ray Diffraction Measurement

Each powder mentioned above was measured by X-ray diffraction. The results are shown below:

1)  $\text{Sr}_3\text{NiPtO}_6$  (Baking Temperature: 500°C)

After the bake, no crystal structure of  $\text{Sr}_3\text{NiPtO}_6$  was detected. More specifically,  $\text{Sr}_3\text{NiPtO}_6$  was not produced through the baking at 500°C.

2)  $\text{Sr}_3\text{NiPtO}_6$  (Baking Temperature: 1000°C)

After the bake, a crystal structure of  $\text{Sr}_3\text{NiPtO}_6$  was detected. However, no crystal structure of  $\text{Sr}_3\text{NiPtO}_6$  was detected after the reduction treatment and the oxidation treatment. More specifically, it seemed that the  $\text{Sr}_3\text{NiPtO}_6$  was disintegrated after the reduction treatment and the oxidation treatment.

3)  $\text{Sr}_4\text{PtO}_6$  (Baking Temperature: 500°C)

After the bake, no crystal structure of  $\text{Sr}_4\text{PtO}_6$  was detected. More specifically,  $\text{Sr}_4\text{PtO}_6$  was not produced through the baking at 500°C.

4)  $\text{Sr}_4\text{PtO}_6$  (Baking Temperature: 1000°C)

After the bake, a crystal structure of  $\text{Sr}_4\text{PtO}_6$  was detected. However, no crystal structure of  $\text{Sr}_4\text{PtO}_6$  was detected after the reduction treatment and the oxidation treatment. More

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specifically, it seemed that the  $\text{Sr}_4\text{PtO}_6$  was disintegrated after the reduction treatment and the oxidation treatment.

5)  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_3\text{NiPtO}_6$  (Baking Temperature: 500°C)

After the bake, no crystal structure of  $\text{Sr}_3\text{NiPtO}_6$  in  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_3\text{NiPtO}_6$  was detected. More specifically, the  $\text{Sr}_3\text{NiPtO}_6$  was not produced in  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_3\text{NiPtO}_6$  through the baking at 500°C.

6)  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_3\text{NiPtO}_6$  (Baking Temperature: 1000°C)

After the bake, no crystal structure of  $\text{Sr}_3\text{NiPtO}_6$  in  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_3\text{NiPtO}_6$  was detected. More specifically,  $\text{Sr}_3\text{NiPtO}_6$  was not produced in  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_3\text{NiPtO}_6$  through the baking even at 1000°C.

7)  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_4\text{PtO}_6$  (Baking Temperature: 500°C)

After the bake, no crystal structure of  $\text{Sr}_4\text{PtO}_6$  in  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_4\text{PtO}_6$  was detected. More specifically, the  $\text{Sr}_4\text{PtO}_6$  was not produced in  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_4\text{PtO}_6$  through the baking at 500°C.

8)  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_4\text{PtO}_6$  (Baking Temperature: 1000°C)

After the bake, no crystal structure of  $\text{Sr}_4\text{PtO}_6$  in  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_4\text{PtO}_6$  was detected. More specifically, the  $\text{Sr}_4\text{PtO}_6$  was not produced in  $\text{MgAl}_2\text{O}_4$  supporting  $\text{Sr}_4\text{PtO}_6$  through the baking even at 1000°C.

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I, the undersigned, declare that all statement made herein on my knowledge are true and that all statements made on information and belief are believed to be true: and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and such willful false statements may jeopardize the validity of the application or any issuing thereon.

Signed this                    day of October, 2007.

*19. October, 2007.*

Kimiyoshi KANEKO

Kimiyoshi KANEKO